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ORIGINAL ARTICLE

Ultrasonic supported oil extraction, process modeling, and optimization by response surface methodology tool from *Croton Macrostachyus leaf*

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Abstract
The present study, ultrasound-assisted extraction (UAE) was used to extract *Croton macrostachyus* leaf oil, which enhances the rate of reaction and temperature in a shorter length of time while also ensuring the highest oil recovery. Through response surface methodology (RSM), the influence of sonication temperature (35–45 °C), sonication time (25–35 min), ultrasound power (250–550 W), and percentage of solid-to-solvent ratio (10–20%) on the oil yield was investigated. The optimal conditions of *Croton macrostachyus* leaf oil were found to be 42 °C of temperature, 30.61 min of sonication time, 425.79 W of ultrasound power, and 18% (w/v) of solid to solvent. Under these circumstances, the optimum extraction efficiency of oil was found as 38.63% with a desirability value of 1.00. In general, these findings suggest that *Croton macrostachyus* leaf can be explored as a promising alternative source of oil and new biodiesel feedstock.

Keywords *Croton macrostachyus* leaf · Oil extraction · Response surface methodology · Optimization · Ultrasound-assisted

1 Introduction

Croton macrostachyus is a semi-deciduous land plant of the Euphorbiaceae family that contains approximately 1,300 tree species, herbaceous plants, and herbs common in the tropical and subtropical regions of the world [17]. It is broadly found in some Eastern African countries such as Eritrea, Ethiopia, Kenya, Tanzania, and Uganda [12]. *Croton macrostachyus* is one of the eight species of Croton that can be widely found in Ethiopia (Meresa et al., 2019). Preliminary phytochemical screening of *Croton macrostachyus* leaf showed the existence of secondary metabolites such as phenolic compounds, tannins, terpenoids, alkaloids, and saponins. Almost all plants from root to leaf are utilized in conventional medicine to cure a various complaints [27]. The leaf of *Croton macrostachyus* has medicinal value (treatment for cough and wounds) (Dubale, Amare Aregahegn, Chandravanshi, Bhagwan Singh Gebremariam, 2015). Oil extraction from *Croton gratissimus* seeds has recently been identified as an appropriate raw material for large-scale biodiesel production [5]. Compared to *Croton macrostachyus* seed, the research on *Croton macrostachyus* leaf is limited or no information about oil extraction from *Croton macrostachyus* leaf. In addition, no reports have been published examining various ultrasonic extraction conditions to obtain oil from *Croton macrostachyus* leaves. This prompted the current research, which aimed to collect experimental data on oil extraction to boost the value of this African crop.

Conventional methods require many organic solvents, high extraction temperatures, and prolonged extraction time, which leads to the breakdown of unsaturated fatty acids, lowering quality of oil [36]. There are numerous

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advanced technologies for extracting oil from the seeds and leaves of the plants [34]. When compared to conventional methods, these technologies are often preferred because of their simplified processes, shorter processing time, and better oil recovery [37]. Nowadays, various modern extraction techniques had been shifted from the traditional solvent to nonconventional methods which include supercritical fluid, enzymatic, microwave-assisted, and ultrasound-assisted extraction (UAE) and for the extraction of energetic additive and oil from seed and leaf of the plant [26]. Comparatively, UAE seems to be a viable technique due to its shorter extraction times, low energy demands, much easier in practice, and high extraction efficiency [31, 38]. The ultrasonic power's intensity causes additional vibration in the sample particles, allowing the solvent to penetrate deeper into the tissue and aiding the recovery of the target component from the solid material to the liquid solvent phase [40].

The impact of ultrasonic extraction process parameters (sonication temperature, sonication time, ultrasound power, and solid-to-solvent ratio) on the extraction of oil from different plant sources was studied by earlier scholars [16, 20]. However, as far as we know, there was no information regarding oil extraction from *Croton macrostachyus* leaf by using ultrasonic-assisted and other separation technologies. Therefore, the objectives of the current study were to scrutinize the impact of extraction conditions (sonication temperature, sonication time, ultrasound power, and percentage of solid-to-solvent ratio) on the recovery of oil from *Croton macrostachyus* leaf as well as to optimize the process parameters using response surface methodology (RSM). Furthermore, the physicochemical properties of the recovered oil under optimal conditions were investigated to evaluate its suitability for the potential relevance to the quality assessments of oil.

2 Materials and methods

2.1 Sample preparation

Based on the weather pattern prevailing in the sampling site, the sampling was done throughout the months of January to April, as these are the months that produce a bumper crop of *Croton macrostachyus* Leaf. The *Croton macrostachyus* leaf used in this investigation was collected randomly at varying stages of maturation (oldest trees, medium trees and youngest trees) from the Jimma Institute of Technology. Afterward, they had been packed in polythene bags and transported to the chemical engineering laboratory at Jimma Institute of Technology. The collected *Croton macrostachyus* leaf was cleaned and air-dried for 4 days until it reached a constant weight. The air-dried *Croton macrostachyus* leaf was ground into powdered form by using a mortar and pestle to increase the surface area for recovery of oil and screened to pass through a 60-mm mesh sieve to achieve the uniform particles. The powdered particles were then sealed in glass bottles and kept at 4 °C for further analysis and recovery of oil.

2.2 Proximate composition of Croton macrostachyus leaf

The proximate composition of *Croton macrostachyus* leaf was determined according to the Standard Official Methods of AOAC (2000). The extraction was carried out in duplicate, and average of two individual was considered for further analysis. The total carbohydrate content was determined according to the following formula (Eq. 1):

$$Total\ carbohydrate(\%) = 100 - (protein(\%) + ash(\%) + fat(\%) + moisture(\%) + fiber(\%)) \quad (1)$$

2.3 Experimental designs and statistics analysis

Statistical experimental design version 11.1.2.06 (STAT-EASE Inc., Minneapolis, USA) was applied to investigate RSM experimental data and generate a three-dimensional (3D) surface. In the present study, the influence of UAE variables and their ranges that were considered for the extraction of oil were sonication temperature (35–45 °C), sonication time (25–35 min), and ultrasound power (250–550 W), and the percentage of solid to a solvent ratio (10–20%, w/v) (Table 3) on the oil yield of *Croton macrostachyus* leaf was investigated. The independent variables and their ranges were selected by considering the previous related works [25, 31]. A three-level four factorial central composite design (CCD) was employed, and the experimental design generates 30 experimental runs [2] by involving 16 factorial points, 8 axial points, and 6 at the center points (Eq. 2).

$$n = 2^k + 2k + m = 2^4 + 2 * 4 + 6 = 30 \quad (2)$$

where n is the total experimental required, k is the number of input variables, and m is the number of central points. The quadratic response model was used to correlate the response to the independent variables (Eq. 3).

$$Y = b_o + \sum_{i=1}^{4} b_i x_i + \sum_{i=1}^{4} b_{ii} x_i^2 + \sum_{i=1}^{3} \sum_{j=i+1}^{4} b_{ij} x_i x_j \quad (3)$$

where Y is the dependent variables, n is the number of process variables, b_0 is the intercept value, b_i is the first-order coefficients, b_{ij} is the interaction coefficients, b_{ii} is the quadratic coefficients, and x_i and x_j are the independent variables.

The analysis of variance (ANOVA) and significance test were employed to analyze the quality of the developed regression equation. The adequacy of the quadratic polynomial model was investigated using the coefficient of determination R^2, predicted R^2, adjusted R^2, probability value and Fisher's test, and lack of fit [2].

2.4 Extraction of oil from Croton macrostachyus leaf

In this study, extraction of oil from *Croton macrostachyus* leaf was conducted according to the methodology proposed by Li et al. [20] with a slight improvement. The *Croton macrostachyus* leaf powder (50 g) was blended with hexane in an Erlenmeyer flask with plugs and sonicated in an ultrasonic water bath (Power sonic 420; frequency 40 kHz; maximum output, 700 W; internal dimensions, 500 × 300 × 150 mm). Upon extraction, the aqueous oil mixture was cooled to room temperature and then centrifuged at a rate of 10,000 rpm for 15 min at 4 °C to separate the oil from the aqueous layer. The supernatant was filtered through Whatman filter paper under a vacuum filter, and the filtrate was separated from solid residues. The solid filtrates were washed twice with the solvent, and a rotary vacuum evaporator was used to a concentrated supernatant solution at 60 °C under vacuum. The oil extracted was calculated and well-kept in a refrigerator at 4 °C for further analysis. The acquired oil was determined according to the weight of oil extracted from *Croton macrostachyus* leaf to the initial weight of leaf powder (50 g). The percentage of oil yield was calculated using the following (Eq. 4) [7].

$$Oil\,yield\,(\%) = \frac{Weight\ of\ the\ oil\ extracts\ (g)}{Initial\ amount\ of\ leave\ powder\ (50g)} x\ 100\% \quad (4)$$

2.5 Fatty acid composition analysis

The composition of fatty acids in *Croton macrostachyus* leaf oil were prepared according to the standard method (AOAC 996.01). The fatty acid composition of each oil extracted from *Croton macrostachyus* leaf was determined by using gas chromatography-mass spectrometry (GC–MS) using an Agilent Technologies 7820A, G4350 GC System equipped with a flame ionization detector (Agilent Technologies Inc., Wilmington, DE, USA), DB-23 capillary column (60 m × 0.25 mm × 0.25 µm), and ChemStation software (Agilent Technologies Inc., Wilmington, DE, USA). The temperature of the column was programmed to rise at a rate of 4 °C/min from 150 to 240 °C at 4 °C/min. The flow rate of the carrier gas, helium, was set at 1 ml/min, and the detector and injector temperatures were set to 240 °C. With a split flow rate of 40 mL/min, the sample injection volume was 2 µL. By comparing the retention periods of the samples to those of the standards, the fatty acids were identified and expressed as milligrams per 100 g of fatty acids.

2.6 Physicochemical characteristics of Croton macrostachyus leaf oil

Chemical analysis of *Croton macrostachyus* leaf oil was carried out at the optimal conditions. The iodine value (IV), acid value (AV), p-anisidine value (PAV), free fatty acid (FFA) contents, saponification value (SV), peroxide value (PV), and specific gravity of *Croton macrostachyus* leaf oil was determined based on the standard method recommended by American Oil Chemists Society (AOCS, 2009) (Table 1).

2.7 Oil stability test

The oxidative stability analysis test of *Croton macrostachyus* leaf oil was evaluated in triplicate for each sample using PV and PAV. Totox value (TV) can be determined using the following (Eq. 5) [31].

$$Y = b_o + \sum_{i=1}^{4} b_i x_i + \sum_{i=l}^{4} b_{ii} x_i^2 + \sum_{i=l}^{3} \sum_{j=i+l}^{4} b_{ij} x_i x_j \quad (5)$$

Table 1 Standard methods used for physicochemical properties of *Croton macrostachyus* leaf oil

Parameter standard	Parameter standard
Acid value (AV), mg/KOH	AOCS Cd 3d-40 (1993) method
Refractive index (RI)	AOCS Cc 7–25 method
Saponification value (SV)	A.O.A.C, (1990) method
Peroxide value (PV), meqO_2/kg oil	AOCS Cd 8–53 (1998b) method
Free fatty acid (% FFA) content	AOAC 996.01 (GLC)
Density (g/cm^3)	AOCS Cc 10a-25(1993) method
Iodine value (IV), meq/kg	AOCS, Cd 1b-87 (1995) method
P-anisidine value (PAV)	AOCS Cd-18–90 (AOCS, 1998a)

Table 2 Proximate compositions of *Croton macrostachyus* leaf (g/100 g dry)

Physical properties (%)	*Croton macrostachyus* leaf	*Croton zambesicus* leaf	*Croton zambesicus* seed
Moisture content	6.79 ± 0.03	7.88 ± 0.03	7.94 ± 0.02
Ash content	6.84 ± 0.00	9.28 ± 0.00	4.15 ± 0.01
Crude protein content	12.56 ± 0.13	13.13 ± 0.12	9.64 ± 0.14
Crude fiber content	15.78 ± 0.01	16.63 ± 0.34	15.45 ± 0.21
Crude fat content	21.84 ± 0.00	15.42 ± 1.49	26.73 ± 0.32
Carbohydrate content	36.19 ± 0.04	37.66 ± 0.32	36.09 ± 2.28
References	Measured	[6]	

3 Result and discussion

3.1 Proximate composition of the Croton macrostachyus leaf

The proximate composition of *Croton macrostachyus* leaf has been shown in Table 2. The ash content of *Croton macrostachyus* leaf obtained was 6.84%, which were higher than the values of *Croton zambesicus* seed (Table 2), sesame seed (5.36%) [11], and date palm seed oil (1.13%) [30] but lower than that of *Croton zambesicus* leaf (9.28%). The moisture content of leaf of *Croton macrostachyus* was higher than that of a sesame seed (5.10) [11] but lower than that of *Croton zambesicus* leaf and seed (Table 2) and date palm seed (9.78%) [30]. The protein content of the *Croton macrostachyus* leaf was 12.56%, which was lower than that of sesame seed (25.73%) [11] and *Croton zambesicus* leaf but higher than *Croton zambesicus* seed (Table 2).

The crude fiber content of *Croton macrostachyus* leaf was slightly lower than that of *Croton zambesicus* leaf but higher than sesame seed (3.42%) [11] and *Croton zambesicus* seed (Table 2). The fat value of *Croton macrostachyus* leaf was 21.84%, low compared to *Croton zambesicus* seed and sesame seed [11] but higher than that of *Croton zambesicus* leaf (Table 2). The amount of carbohydrates found in the leaf of *Croton macrostachyus* was 36.19%, which was similar to that of the leaf and seed of *Croton macrostachyus* (Table 2) but higher than sesame seed [11]. The result reveals that the variation of the proximate composition of Croton might be influenced by the variety of croton plants, cultivation climate, ripening stage, soil type, harvesting time of the leaf, and the extraction technique used. In general, the results of proximate composition of *croton macrostachyus* leaf were consistent with the results described in the literature (Table 2).

3.2 Statistical analysis of the regression model for oil extraction

Statistical experimental design was applied to assess the experimental and predicted oil yields. The 30 experimental runs had been carried out with various interactions of process factors and the actual yields were compared with the predicted ones as shown in Table 3. By comparing the magnitude of the factor coefficients, the coded equation can be used to determine the significant effects of the process variables. The developed quadratic model equation that correlates the oil yield with the coded four process variables (sonication temperature (A), sonication time (B), ultrasound power (C), and the ratio of solid to the solvent (D)) after eliminating the insignificant variables were given in Eq. (6). The developed equation was valid within the range of examined conditions: 30 °C < sonication temperature < 50 °C, 20 min < sonication time < 40 min, 100 W < ultrasound power < 700 W, and 5% < ratio of solid to the solvent < 25%.

$$Oilyeild(\%) = 38.01 + 3.24A + 1.29B + 0.59C + 0.9737D + 1.69AC + 1.56AD - 1.30BC - 1.26CD - 7.72A^2 - 2.99B^2 - 1.91C^2 - 1.67D^2 \quad (6)$$

The negative coefficients describe that the factors negatively affect the yield of oil, whereas the positive coefficients show that the factors positively affect the yield of oil.

The second-order polynomial equation was estimated statistically using ANOVA, *F* test, and *p* value, and the results were displayed in Table 4. The ANOVA results revealed that the quadratic regression model was a perfect match (*F* value of 94.47) ($p < 0.0001$, indicate that the model was significant at the confidence level of 95% ($p < 0.05$)) (Table 4). The lowest *p* value and the largest *F* value indicate that the model was the most substantial ($p < 0.05$) effect on the corresponding response [3, 18]. The values of lack of fit 3.62 and 0.084 acquired for *F* value and *p* value, respectively, indicate that the model was insignificant. Thus, the results obtained verified that the mentioned models (Eq. 6) were sufficiently accurate in terms of predicting the response within the selected range of variables examined. The response acquired from Table 4 shows that all the linear coefficients (A, B, C, D), interaction term (AC, AD, BC, CD), and the quadratic term (A^2, B^2, C^2, D^2) had notable effect on oil yield, while AB and BD were insignificant effects on oil yield.

The accuracy of the model was verified by determining the value of R^2, adj R^2, predicted R^2, Adeq Precision, and variation coefficient (CV). Table 5 shows that the summary

Table 3 Experimental yields and composite design matrix runs

Run	Coded variable				Decoded variable				Dependent variable	
	A	B	C	D	Temp (°C)	Time (min)	Power (W)	% Solid to solvent	Actual oil yield (%)	Predicted oil yield (%)
1	1	−1	1	1	45	25	550	20	30.38	30.32
2	1	−1	−1	1	45	25	250	20	25.68	25.69
3	1	1	−1	−1	45	35	250	10	23.64	24.32
4	−α	0	0	0	30	30	400	15	3.09	0.6621
5	1	−1	−1	−1	45	25	250	10	18.68	17.45
6	−1	1	−1	−1	35	35	250	10	22.34	23.29
7	0	0	α	0	40	30	700	15	32.56	31.57
8	0	α	0	0	40	40	400	15	29.68	28.64
9	0	0	0	0	40	30	400	15	36.46	38.01
10	−1	−1	1	1	35	25	550	20	18.5	18.40
11	α	0	0	0	50	30	400	15	12.65	13.61
12	−1	−1	1	−1	35	25	550	10	20.63	21.46
13	0	0	0	0	40	30	400	15	38.46	38.01
14	0	-α	0	0	40	20	400	15	23.92	23.49
15	−1	−1	−1	−1	35	25	250	10	17.48	18.55
16	−1	1	1	-1	35	35	550	10	20.45	21.02
17	−1	1	−1	1	35	35	250	20	24.18	24.01
18	0	0	0	−α	40	30	400	5	30.38	29.38
19	0	0	-α	0	40	30	100	15	29.68	29.21
20	1	−1	1	−1	45	25	550	10	26.39	27.14
21	0	0	0	0	40	30	400	15	38.39	38.01
22	−1	1	1	1	35	35	550	20	14.56	16.68
23	0	0	0	0	40	30	400	15	37.98	38.01
24	0	0	0	α	40	30	400	25	33.75	33.28
25	1	1	1	1	45	35	550	20	31.22	30.73
26	0	0	0	0	40	30	400	15	38.39	38.01
27	1	1	11	−1	45	35	550	10	28.98	28.82
28	0	0	0	0	40	30	400	15	38.39	38.01
29	1	1	−1	1	45	35	250	20	31.22	31.28
30	−1	−1	−1	1	35	25	250	20	19.48	20.54

statistics of the developed regression equation model. The lower value of CV (4.86%) indicates that the variation between the predicted and experimental values was low [35].

The reproducibility measurement was determined by CV, and a value less than 10 is required for the model to be considered more accurate and reliable [28]. The R^2 value of 0.9888 acquired from the model indicates that more than 98.88% of all data set were in agreement with the experimental values. According to Aklilu [3], adjusted R^2 and predicted R^2 must be within 20% to be in reasonable agreement. The predicted R^2 of 0.9413 was in reasonable agreement with the adjusted R^2 of 0.9783, indicating that the requirement was met in this study. A value greater than four was required for the measure of signal-to-noise ratio, which was estimated by adequate precision [35]. The value of adequate precision 40.8563 shows that the requirement was satisfied in this study. All these statistical parameters indicate that the quadratic regression equation was highly accurate and reliable for the extraction procedures. The predicted values acquired from the quadratic regression equation were close to the observed values and approximately along a straight line as shown in Fig. 1. Indeed, the high value of R^2 (0.9888) ratifying that the regression model is suitable for predicting oil yield.

3.3 Effect of process variables on the oil recovery

The oil content of *Croton macrostachyus* leaf varied from 3.09 to 38.46% (Table 6), which are comparable to the conventional sources of seed oil content of soybean (15–20%),

Table 4 Analysis of variance for oil extraction from *Croton macrostachyus* leaf

Source	Sum of squares	Df	Mean square	*F* value	*p* value
Model	2210.66	14	157.90	94.47	< 0.0001
A, Temperature	251.49	1	251.49	150.46	< 0.0001
B, Time	39.76	1	39.76	23.79	0.0002
C, Power	8.37	1	8.37	5.01	0.0409
D, % solid	22.76	1	22.76	13.62	0.0022
AB	4.51	1	4.51	2.70	0.1214
AC	45.87	1	45.87	27.44	0.0001
AD	39.03	1	39.03	23.35	0.0002
BC	26.91	1	26.91	16.10	0.0011
BD	1.62	1	1.62	0.9688	0.3406
CD	25.53	1	25.53	15.27	0.0014
A^2	1634.21	1	1634.21	977.74	< 0.0001
B^2	244.62	1	244.62	146.35	< 0.0001
C^2	99.68	1	99.68	59.64	< 0.0001
D^2	76.51	1	76.51	45.77	< 0.0001
Residual	25.07	15	1.67		
Lack of fit	22.03	10	2.20	3.62	0.0840
Pure error	3.04	5	0.6078		
Cor total	2235.73	29			

Table 5 Model summary statistics

S. No	Parameters	Values
1	Standard deviation	1.29
2	Mean	26.59
3	Variation coefficient (CV %)	4.86
4	R^2	0.9888
5	Adjusted R^2	0.9783
6	Predicted R^2	0.9413
7	Adeq Precision	40.8563
8	Model suggested	Quadratic

Pongamia pinnata (27–39%), peony seed (25.35–30.85%), sunflower (25–35%), and *Ceiba pentandra* (25–28%) as shown in Table 6. However, the oil content of *Croton macrostachyus* leaf was lower than *Jatropha curcas* crude oil (55–60%), palm oil (30–60%), *Croton macrostachyus* seeds oil (50–70%), rubber seed oil (53.74–68.35%), *Calophyllum inophyllum* (65–75%), and castor seed oil (45–50%) [15, 20, 33]. The current results revealed that the recovered oil from *Croton macrostachyus* leaf was consistent with the values acquired from the commercially grown seed oil (soybean, sunflower, castor seed, and *Jatropha curcas*), thus demonstrating that the potential utilization of inexpensive raw material as an alternative source for biodiesel production and soap making.

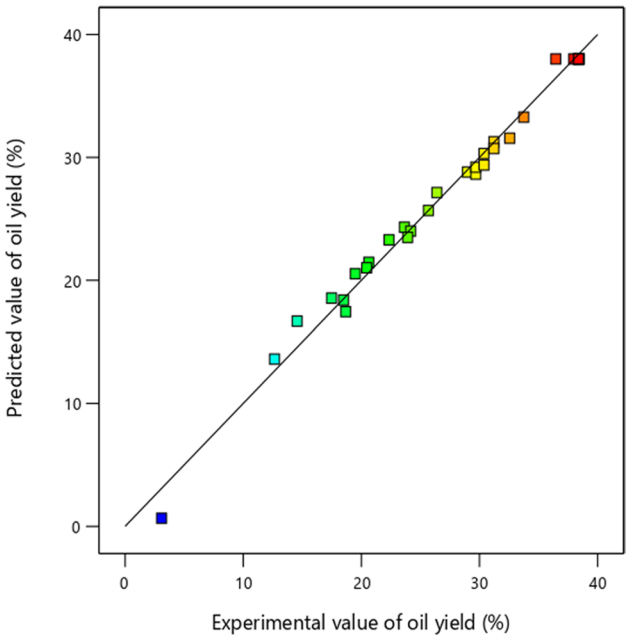

Fig. 1 The relationship between the observed and model-predicted value of oil yield

Table 6 Oil content from different sources

Feedstock types	Oil yield (%)	References
Peanut	45–55	[15]
Ceiba pentandra	25–28	[32]
Sunflower	25–35	[25]
Pongamia pinnata	27–39	[33]
Rapeseed	38–46	[25]
Soybean	15–20	[15]
Linseed	40–44	[26]
Canarium odontophyllum kernel	52.24–63.27	[26]
Coconut	63–65	[15]
Croton macrostachyus seeds	50–70	(Aga et al., 2020)
Palm oil	30–60	[15]
Rubber seed	53.74–68.35	[33]
Calophyllum inophyllum	65–75	[32]
Jatropha curcas	55–60	[32]
Microalgae	30–70	[15]
Castor	45–50	[15]
Canola	40–45	[15]
Peony seed	25.35–30.85%	[20]
Current study	3.09–38.46%	

Figures 2a, b, c, and d depict the influence of an independent variable on the recovery of oil. Percent of oil recovery was directly proportional to sonication temperature, sonication time, ultrasound power, and percentage of solid-to-solvent ratio, implying that increasing any of those

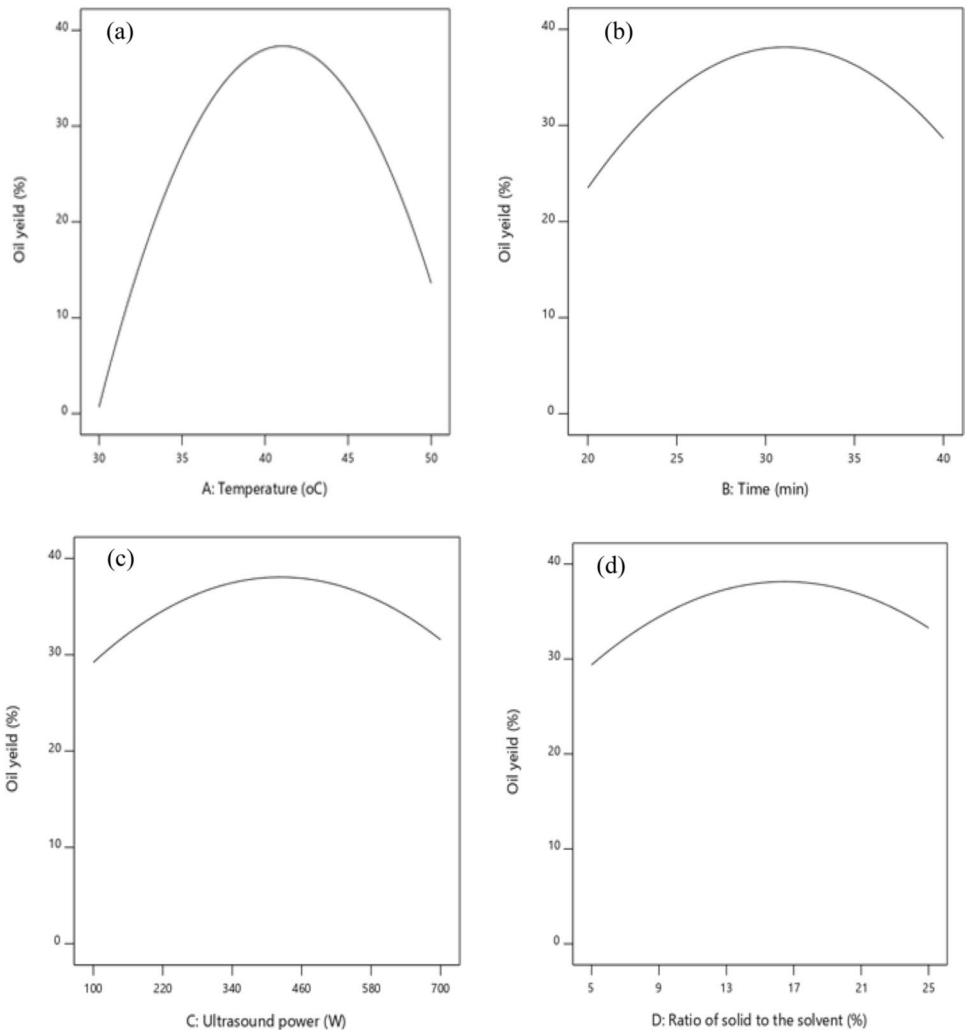

Fig. 2 Effect of independent variables: **a** sonication temperature, **b** sonication time, **c** ultrasound power, and **d** percentage of solid-to-solvent ratio on the oil yield

independent variables would enhance oil extraction until the optimal values were achieved (Eq. 6). However, increasing those process variables further will result in a considerable reduction in oil extraction since too high those values result in oil decomposition and reduced solvent volume.

The influence of sonication temperature on the oil extraction was shown in Fig. 2a, which displays that oil extraction increases with sonication temperature increasing while holding all other factors constant. The results revealed that sonication temperature has a substantial influence on the recovery of oil, followed by sonication time, percentage solid-to-solvent ratio, and ultrasound power (Table 4). The sonication temperature was directly proportional to the recovery of oil, and it has been shown that increasing the value of sonication temperature would increase the oil extraction (Eq. 6). From Table 3, it was observed that the oil yield at 40 and 45 °C was considerably higher than that at 25, 30, and 35 °C, which was probably due to enhanced mass transfer of oil from the leaf powder into the extraction medium. This phenomenon was agreed well with the optimization of peony seed oil as reported by Li et al. [20]. Increasing the temperature in the UAE may improve oil extraction efficiency by creating more air bubbles and increasing the contact surface between solvent and cells' membranes according to various studies [16, 21]. Nevertheless, when the sonication temperature exceeded 45 °C, the oil yield decreases due to the rapid evaporation of the solvent at high temperature and decrease in the viscosity of the solvent. This phenomenon was agreed well with Tan et al. [36], who reported that the yield of virgin avocado oil increased as the temperature increased from 20 to 40 °C and canola seed oil with Jalili et al. [16], who reported that the recovery of oil boosted from 18 to 20% when the applied temperature was increased by 40 to 50 °C.

The impact of sonication time on the extraction of oil was shown in Fig. 2b, which elucidates that the extraction of oil increased as sonication time increased while maintaining all other variables unchanged. Sonication time was directly proportional to the oil yield and suggests that increasing the value of time favored the extraction of oil until the maximum

value was reached (Eq. 6). Due to the prolonged sonication time increasing the cavitation phenomenon, contact time of solvent with leaf powder, so that severe cell wall crushing occurs, and the interaction surface between solvent and oil cells increased, leading to increased oil recovery. However, when the oil yield reaches the optimum value, it begins to gradually decline as the sonication time increases due to the decomposition of the oil. A similar phenomenon was also observed in UAE of peony seed oil optimization [20].

The influence of ultrasound power on oil extraction was illustrated in Fig. 2c, which indicates that the oil extracted from the leaf of *Croton macrostachyus* increased as the ultrasound power increased if all other variables unchanged (Eq. 6). When ultrasound power increased, the oil yield increased initially until the optimum value was reached, but start to decrease when ultrasound power exceeds 450 W. This is might be due to the ultrasound power altering the structure of plant tissues and thermal decomposition of volatiles and increasing the solvent permeability in the plant matrix. In addition to this, increasing ultrasound power accelerates the diffusion ratio to a certain value, thereby improving extraction of oil from *Croton macrostachyus* leaf. A similar phenomenon was presented in the extraction of oil from *Calophyllum inophyllum* seeds by the UAE [29].

The influence of percent solid-to-solvent ratio on oil yield was varying in ranges from 5 to 25% was examined by keeping other variables unchanged. Compared to solid-to-solvent ratio of 15% and 20%, 5%, 10%, and 25% produced lower oil yield, but the difference among these solid-to-solvent ratios was not significant (Table 3). Figure 2d depicts the recovery of oil increased as the % solid-to-solvent ratio increased by maintaining all other factors unchanged. The percentage of solid-to-solvent ratio has a direct relationship with oil yield, and it has been shown that increasing percentage of solid-to-solvent ratio leads to an increase in oil yield since the oil concentration gradient between solvent and solid becomes higher, which promotes good mass transfer and leads to more efficient extraction. However, the oil yield decreased when the ratios exceed 20%, this is might be due to the decomposition, saturating solvent recovery, and volatilization of oil in the distillation. A similar phenomenon has

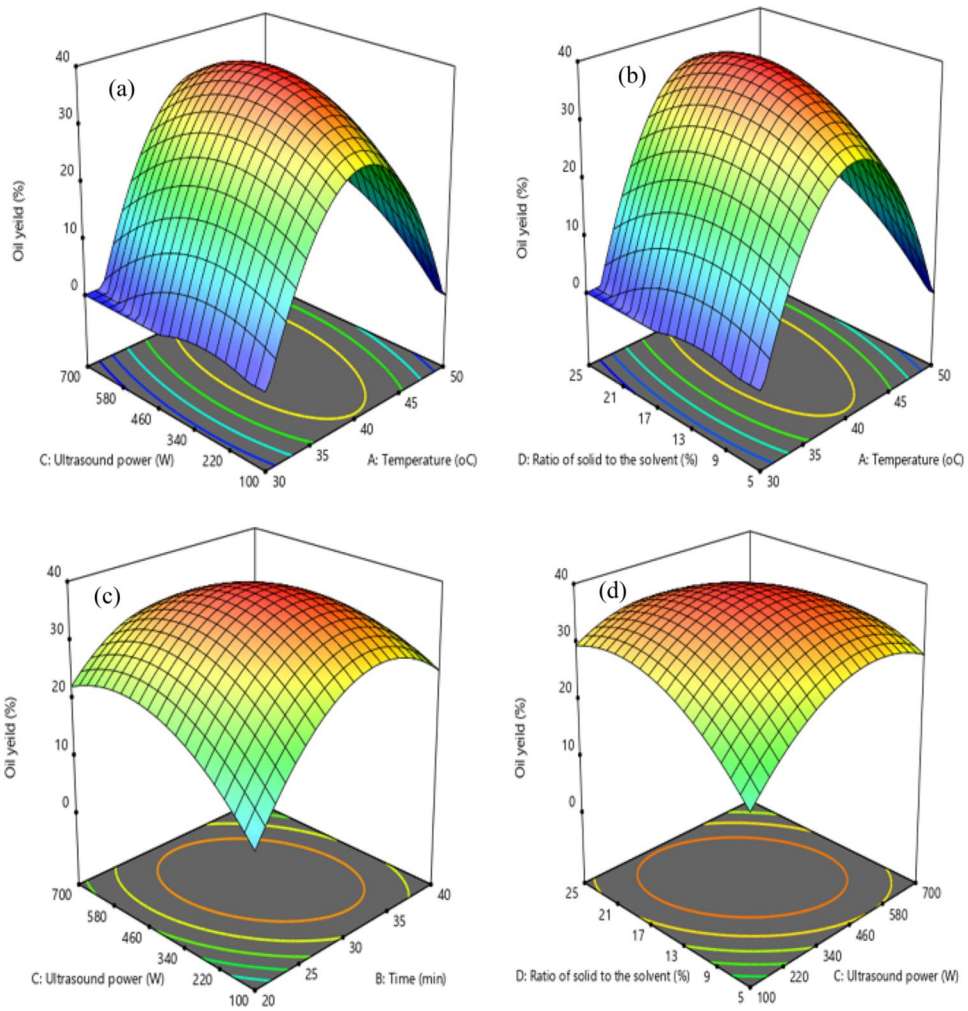

Fig. 3 The three-dimensional surface plot of oil yield: **a** the influence of sonication temperature and ultrasound power at solid-to-solvent ratio of 15% and sonication time of 30 min; **b** effects of sonication temperature and percentage of solid-to-solvent ratio at sonication time of 30 min and ultrasound power of 400 W; **c** effects of ultrasound power and sonication time at sonication temperature of 40 °C and solid-to-solvent ratio of 15%; **d** effects of the percentage of solid-to-solvent ratio and ultrasound power at sonication temperature 40 °C and sonication time of 30 min

been presented in annatto seeds [38], peony seed oil [20], and papaya seed oil [31].

3.4 The effects of process variables interaction on the oil yield

The influence of interaction between independent process variables on oil recovery was scrutinized using response surface plots as shown in Fig. 3a, b, c, and d. The result revealed that the interaction of sonication temperature and ultrasound power has the most substantial ($p < 0.0001$) influence on the oil yield, followed by sonication temperature with a percentage of solid to solvent ratio, sonication time with ultrasound power, and ultrasound power with a percentage of solid-to-solvent ratio (Table 4).

Figure 3a shows the impact of the interaction between sonication temperature and ultrasound power, while sonication time and percentage of solid-to-solvent ratio held at the constant level. In this surface plot, the percentage of oil recovery increased rapidly with increasing the effect of interaction between sonication temperature and ultrasound power up to an optimum point. However, oil yield slightly decreases as the interaction between sonication temperature and ultrasound power increased since higher sonication temperature causes degradation of oil composition.

Figure 3b elucidates the impact of the interaction between sonication temperature and the % of solid-to-solvent ratio on the extraction of oil at constant sonication time and ultrasound power. When compared with sonication temperature and solid-to-solvent ratio at constant ultrasound power and sonication time, the impact of sonication temperature was more prominent on oil yield because the system had already attained equilibrium at high temperature. Increasing the interaction between temperatures with the % ratio of solid to solvent increased the extraction of oil. However, a further increment of both variables exhibited a decreasing tendency in oil yield. A similar phenomenon was observed during oil extraction from *Diospyros lotus* seed [34].

The combined impact of sonication time and ultrasound power on oil yield has been presented in Fig. 3c. This surface plot reveals that the oil yield decreases as sonication time and ultrasound power increase up to an optimum point, but the sonication time had a more prominent impact than ultrasound power (Table 4). In fact, in order to increase the contact surface between oil cells and solvent, an appropriate sonication time was required to induce the diffusion of hexane in the extraction medium. The surface plot reveals that the effect of sonication time prevailed over the effect of ultrasound power, as previously reported [15].

Figure 3d represents the response surface plot showing the interaction of ultrasound power with a ratio of solid to the solvent for oil yield. From the surface plot, the oil recovery increased as both ultrasound power and the ratio of solid to solvent decreased. In addition to this, the influence of the percentage of solid-to-solvent ratio was more powerful when compared with ultrasound power at constant sonication temperature and time. The interaction between the percentage of solid-to-solvent ratio and ultrasound power demonstrated a less impact on oil yield compared to other combined factors (Table 4).

3.5 Optimization and verification of oil yield model

The validation of the predictive regression equation was performed under the optimal conditions of the variables of the extraction process. The model validation for oil yield is shown in Table 7. An optimum condition for the response was set at the maximum value, while the independent variables were set at the values within the study range. Triplicate extractions were performed under optimum conditions to compare the actual values with the predicted values using developed regression (Eq. 6). The optimal conditions of *Croton macrostachyus* leaf oil were found to be 42 °C of temperature, 30.61 min of sonication time, 425.79 W of ultrasound power, and 18% of solid-to-solvent ratio. Under these conditions, the optimum oil yield was found as 38.63% with a desirability value of 1.00. The slight variation (< 2.22%) between the actual and the predicted response confirmed the accuracy of the estimating models developed through statistical experimental design. The model's validity was established, and the presence of the optimal point was confirmed by the close agreement between the predicted and experimental findings.

3.6 Physicochemical properties of Croton macrostachyus leaf oil

Physicochemical characterization of *Croton macrostachyus* leaf oil was carried out under optimal operating conditions to ensure the oil quality. Table 8 shows that the physic-chemical analyses of *Croton macrostachyus* leaf oil.

Croton macrostachyus leaf oil had a refractive index (RI) of 1.473 (Table 8), slightly higher than FAO/WHO recommended values, but within a range comparable to the values in the literature. With an increase in the iodine value of the oil, the RI seemed to rise. This could suggest that the higher the RI, the more unsaturation of fatty acid content and the optical characteristic of the oil. This result was consistent with a RI of *croton gratissimus* oil (1.472) [5], *Ricinus*

Table 7 Results of statistical analysis for confirmation of optimization

Response	Predicted value	Actual value	Desirability	Error
Oil yield (%)	38.63	37.79	1.00	2.22

Table 8 The physicochemical characterization of *Croton macrostachyus* leaves oil

Characteristic parameter	Current value (%)	FAO/WHO standards	Literature value
The density of oil at 25 °C, (kg/m^3)	914	870	870–930
kinematic viscosity (mm^2/s)	33.65	39.7	45–222
Refractive index	1.473	1.463	1.458–1.487
Saponification value (mg KOH/g)	185.79	181.40	96.4–199
Iodine value (ml/g)	95.98	80–106	64.1–79.95
Acid value (mg KOH/g oil)	4.18	4	2.4–5.6
Free fatty acids (%FFA)	1.74	0.57–0.728	0.33–1.7
Peroxide value (g/100 g)	2.6		0.5–7.6
Totox value (meq/kg)	9.76		
P-anisidine value (meq/kg)	4.56		

communis seed oils (1.46–1.79), *Hiptage benghalensis* seed oil (1.474), and *Jatropha curcas* (1.46) [10].

The present of FFA in the fat as well as in oil was measured by acid value (AV) to monitor oil degradation during storage [23]. The oil extracted from *Croton macrostachyus* leaf had an AV of 4.18 mg KOH/g (Table 8), which is consistent to oil extracted from papaya seed oil (1.26 mg KOH/g) and higher than peony seed oil (2.20) [23] but lower than high AV of three oils such as *Jatropha curcas* oil (25.4 mg KOH/g), *Calophyllum inophyllum* oil (46.6 mg KOH/g), and *Ceiba pentandra* oil (33.6 mg KOH/g) [32].

Oil extracted from *Croton macrostachyus* leaf had a kinematic viscosity of 33.65 mm^2/s (Table 8), which was higher than that of *Jatropha curcas* oil (28.35 mm^2/s) but lower than *Calophyllum inophyllum* oil (53.17 mm^2/s) and *Ceiba pentandra oil* (34.45 mm^2/s) [32]. When compared with the viscosity of most oils currently utilized for biodiesel synthesis, the *Croton macrostachyus* leaf oil has high kinematic viscosity (33.65 mm^2/s). According to Haji-Moradkhani et al. [13], the density of the oil is a suitable criterion for detecting and evaluating the type of oil. The density of the oil extracted from *Croton macrostachyus* leaf was found to be 914 kg/m^3 (Table 8), which was comparable to that of the oil extracted from *Canarium odontophyllum* kernel (913.7 kg/m^3), *Jatropha curcas* (922.1 kg/m^3), and palm kernel (923.4 kg/m^3) [15] while higher than papaya seed (870 kg/m^3) [39].

The iodine value (IV) of oil extracted from *Croton macrostachyus* leaf was 95.98% (Table 8), which falls within the range of FAO/WHO recommendation despite being lower than *Paeonia ostii* seed oil (161.92–204.46 g/100 g) [23] and papaya seed oil (61.73 g/100 g) [39]. The current results were comparable with those obtained from the commercially grown vegetable oils (soybean, peanut, rapeseed, sunflower seed, corn, sesame, safflower seed, walnut, and rice bran oil) [1]. A higher IV indicates the existent of more C=C bonds in the oil [22] and a high content of unsaturated fatty acids in *Croton macrostachyus* leaf.

The oil extracted from leaf of *Croton macrostachyus* had saponification value (SV) of 185.79 mg KOH/g, which was within the ASTM range of values and was comparable to castor seed oil (185.83 mg KOH/g) [4], but lower than *Croton megalocarpus* seed oil (194.9 mg KOH/g) [5] and papaya seed oil (209.07 mg KOH/g) and higher than FAO/WHO standards (80–106 mg KOH/g) [39]. The results of this study were lower than those published by other researchers for the most commercially grown oil sources such as palm oil (200 mg KOH/g), groundnut oil (193 mmerg KOH/g), and coconut oil (257 mg KOH/g) reported by Nchimbi [25]. The %FFAs and SV compositions in the oil are comparable to the recommended standards values for oil quality. Accordingly, the *Croton macrostachyus* leaf oil obtained in this study satisfies the suggested criteria for efficient exploitation of raw materials for the production of good quality soap and biodiesel production.

Oil produced from *Croton macrostachyus* leaf had a p-anisidine value (AV) of 0.69 g/100 g, which was lower than that of papaya seed oil [31]. The PV of oil extracted from *Croton macrostachyus* leaf was found to be 2.6 g/100 g, which was lower than tomato seed oil (17.52 g/100 g) [28] but higher than papaya seed oil (1.03 mg O_2/kg) [39] and *Camellia oleifera* seeds oil (0.04–0.10 g/100 g) [14]. The PV is significantly lower than the published values illustrating the good quality of the oil obtained in the present study, because low PV improves oil stability.

Totox value (TV) of *Croton macrostachyus* leaf oil was found to be 9.76 meq/kg, which was higher than that of papaya seed oil [31]. Thus, by comparing the FFA composition of the *Croton macrostachyus* leaf oil from the present study with other feedstocks used for biodiesel synthesis, it can be concluded that *Croton macrostachyus* leaf oil is a viable and promising feedstock for methyl ester production. Furthermore, the physicochemical properties of *Croton macrostachyus* leaf oil were consistent with those of *Paeonia lactiflora* pall seed oil, peony seed oil, and *Croton*

Table 9 Comparison of fatty acid composition of oil extracted from *Croton macrostachyus* leaf

Fatty acid	*Croton macrostachyus* leaf oil	*Croton gratissimus* seed oil	*Croton zambesicus* oil	
			Leaf	Seed
Carproic acid (C6:0)	0.82	-	-	-
Carproic acid (C8:0)	0.67	-	-	-
Carproic acid (C10:0)	2.54	-	-	-
Lauric acid (C12:0)	1.97	-	1.04	1.10
Myristic acid (C14:0)	1.47	-	1.41	1.50
Palmitic acid (C16:0)	38.91	36.47	32.03	30.72
Stearic acid (C18:0)	11.97	32.12	4.81	4.92
Cis-9 oleic acid (C18:1)	8.48	8.91	33.28	34.14
Linoelaidic acid (C18:2)	12.24	9.63	18.27	18.46
Arachidic acid (C20:0)	1.32	2.34	0.59	0.54
Cis-11-Eicosenoic add (C20:1)	18.62	-	-	-
Lignoceric acid (C24:0)	0.98	-	0.59	0.63
Total polyunsaturated fatty acids	12.24	14.40	21.20	21.36
Total monounsaturated fatty acids	27.1	9.23	36.96	37.79
Total saturated fatty acids	60.65	75.29	41.82	40.85
References		[5]	[6]	

seed, indicating that *Croton macrostachyus* leaf might use as non-edible oil sources.

The oil extracted from *Croton macrostachyus* leaf had a free fatty acids (%FFA) of 1.74 (Table 8), which was higher than the recommended values of FAO/WHO while being lower than water plant (3.7 to 3.9%), palm kernel oil (4.81%), palm oil (5.05%) [25], and *Hiptage benghalensis* seed oil (1.78%) [10]. The concentration of FFAs in the oil is used as a common parameter to evaluate the quality of oil during extraction, storage, and marketing [24]

The fatty acid composition of oil extracted from *Croton macrostachyus* leaf and those of similar feedstock oils were shown in Table 9. In this study, oil extracted from *Croton macrostachyus* leaf had a total unsaturated fatty acid content of 39.34%. The key five dominating total fatty acid compositions were palmitic acid (C16:0), Cis-11-Eicosenoic add (C20:1), linoelaidic acid (C18:2), stearic acid (C18:0), and Cis-9 oleic acid (C18:1). The most abundant fatty acid in the oil extracted using UAE was palmitic acid (38.91%), which was higher than 32.03% palmitic acid in *Croton zambesicus* leaf and lower than palm oil (41.78%) [6]. Total saturated fatty acids were found to be the most prevalent when compared to unsaturated fatty acid composition (Table 9). Lignoceric acid (0.98%), carproic acid (0.67%), lauric acid (1.97%), myristic acid (1.47%), and arachidic acid (1.32) were found in small amounts (Table 9). The total saturated fatty acid content of the examined *Croton macrostachyus* leaf oil was higher than in previous studies of various non-edible oil sources [5, 6, 8, 19, 21]. Furthermore, the presence of saturated fatty acid in this species suggested that might be useful in cosmetics and consumable items.

4 Conclusion

In the present study, the influence of the variables of ultrasound extraction process (sonication temperature, sonication time, ultrasound power, and percentage of solid-to-solvent ratio) on oil yield of *croton macrostachyus* leaf was investigated by using RSM. The study revealed that sonication temperature has the most significant ($p < 0.0001$) effect on the oil yield, followed by sonication time, solid-to-solvent ratio, and ultrasound power. The optimal conditions for *Croton macrostachyus* leaf oil were found to be 42 °C of sonication temperature, 30.61 min of sonication time, 425.79 W of ultrasound power, and 18% (w/v) of solid to solvent. In these conditions, the optimum oil extraction efficiency was found to be 38.63% with a desirability value of 1.00. The results of the physicochemical characteristics of the oil revealed that it can be utilized as a good source of oil and a promising feedstock for biodiesel extraction. In general, the result of the study suggested that the UAE was an effective and viable technique to recover the crude oil from the *Croton macrostachyus* leaf.

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Venkata. The first draft of the manuscript was written by Ermias and all authors commented on previous versions of the manuscript. All authors read and approved the final manuscript.

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Availability of data and materials All data analyzed during this study are included in this research article.

Declarations

Competing of interest The authors declare no competing interests.

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